

# ME FILE CUI.

# **CRL**

## **COLUMBIA UNIVERSITY**

# AD-A198 060

# SPONTANEOUS AND INDUCED COHERENT RADIATION **GENERATED IN ATOMIC VAPOR**

#### FINAL REPORT

1978 - 1988

**TO THE** 

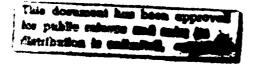
#### OFFICE OF NAVAL RESEARCH

Contract #N00014-78-C-0517



COLUMBIA RADIATION LABORATORY New York, New York 10027

July 1988



REPORT DOCUMENTATION PAGE		READ INSTRUCTIONS BEFORE COMPLETING FORM
REPORT NUMBER	2 COVT ACCESSION NO.	3 RECIPIENT'S CATALOG NUMBER
TITLE (and Subittle)		5 TYPE OF REPORT & PERIOD COVERED
		Final Report
SPONTANEOUS AND INDUCED COHERENT RADIATION		06/01/78-05/31/88
GENERATED IN ATOMIC VAPOR		6 PERFORMING ORG REPORT NUMBER
- AUTHOR(s)		8 CONTRACT OR GRANT NUMBER(#)
PROFESSOR SVEN R. HARTMANN		N00014-78-C-0517
PERFORMING ORGANIZATION NAME AND ADDRESS Columbia University		10. PROGRAM ELEMENT PROJECT, TASK AREA & WORK UNIT HUMBERS
Columbia Radiation Laborate	ory	
538 W. 120th St., New York	, NY 10027	
1 CONTROLLING OFFICE NAME AND ADDRESS		12. REPORT DATE
		07/28/88
		13. NUMBER OF PAGES
MONITORING AGENCY NAME & ADDRES	55(II dillerent from Controlling Office)	15 SECURITY CLASS. (of this report)
Office of Naval Researd		
33 Third Ave. Lower Level		154 DECLASSIFICATION DOWNGRADING
New York, NY 10003-99	998	SCHEDULE
DISTRIBUTION STATEMENT (of this Rep	ortj	<u> </u>
unlimited		
DISTRIBUTION STATEMENT (of the abet	ract entered in Block 20, if different tro	en Report)
unlimited		
SUPPLEMENTARY NOTES		
KEY WORDS (Continue or reverse elde if i	necessary and identify by block numbers	
<pre>Photon Echo, Stimulated Collisionally Induced Re</pre>	elaxation. Attosecond Ro	NOTO * 111 + m - £ + - 1 - 1 - 1 +
Spectroscopy; Diffraction	on Free Beams, Time Dela	ayed Four Wave Mixing
		∠
ABSTRACT (Continue on reverse side if n	ecessary and identify by block number)	
· Onti1		114-1 41

Optical coherent techniques have been developed and applied in order to obtain relaxation and spectroscopic information in atomic vapors. Theoretical work was performed to understand collisional relaxation and the interaction of intense fields with matter. The utility of using incoherent light fields in generating coherent excitations was demonstrated and analyzed. Applications of the time delayed four wave mixing technique using broadband light for investigating relaxation in solids have begun.

DD 1 JAN 73 1473 EDITION OF 1 NOV 65 IS OBSOLETE

S N 0102- LF- 014- 6601

SECURITY CLASSIFICATION OF THIS PAGE (When Date Entered)

### SUMMARY OF WORK SUPPORTED BY ONR 1978 - 1988 (CONTRACT # N00014-78-C-0517)

#### SPONTANEOUS AND INDUCED COHERENT RADIATION GENERATED IN ATOMIC VAPOR

by Sven R. Hartmann, Professor of Physics Columbia University in the City of New York

We have developed and applied various optical coherent techniques in order to study relaxation processes in gases as well as to obtain a better understanding of the interaction between light and matter.

Using the tri-level technique we were able to study the collisionally induced relaxation of Rydberg states in Na vapor to principal quantum numbers, n, as high as 40. These measurements were able to follow the relaxation behavior over the low n regime where the collision cross section increased with increasing n as well as the high n regime where the opposite occurs. 1.7.9.10.34.38 The related Raman Echo was used to study collision broadening of the  $6P_{1/2}$ - $6P_{3/2}$  transition in Tl vapor. 17.20 Another related phenomena is the two-photon excited state tri-level echo. 31.33.37 This echo, of interest in its own right, is useful for studying superposition state relaxation of states unaccessible by ordinary photon echo techniques.

We were able to induce spatial gratings to reform. These novel echoes have signatures relevant to spectroscopic and velocity changing collisions studies.<sup>2,5</sup>

Stimulated echoes were used to study general collisional relaxation effects in gases.<sup>3,4</sup>,

Two-pulse photon echoes were used to provide the first demonstration of the contribution of velocity-changing-like effects to the transverse relaxation of atoms in superposition of two states even when the states follow different post-collision trajectories.<sup>6</sup> Related works allowed us to study foreign gas broadening of the Na and Li D lines.<sup>8,13,16,21</sup> The potential of the two-pulse photon echo technique for performing high resolution spectroscopy was demonstrated in an experiment which showed that echoes could be obtained over a dynamic range exceeding 10<sup>12</sup>.<sup>27</sup>

An extensive theoretical analysis was made of the properties of collision kernels in atomic physics problems.<sup>19</sup>

SPECTED 2

on For |A&I| |} ||ced| |ation

ition/

ail and/or Special

A-1

A diagrammatic technique was developed to aid in the analysis of pulse type optical coherent experiments. This technique was further developed into the practical Billiard Ball Model<sup>24,26</sup> which has provided a simple pictorial method of analyzing generalized echo experiments. The elliptical Billiard Ball Model stimulated experiments which gave added insights into echo behavior and confirmed its validity. 22,23,25,27,28

A theorem stating that optical coherent transients from an excited optically thin sample can produce coherent radiation signals for a time no longer than the length of the total excitation pulse had been proposed, and we pointed out that it was not correct.<sup>14</sup>

We considered and commented upon the spectral modification and temporal modification of a narrow-band light pulse propagating through an atomic absorber that has a still narrower spectral hole.<sup>12</sup>

Another experiment of generalized interest involved the properties of "Diffraction Free Beams." We demonstrated novel properties of these beams involving their inability to be blocked.<sup>40</sup>

We demonstrated that it is possible to generate photon echoes with incoherent light.<sup>29</sup> Before this experiment it was widely held that coherent light was necessary to generate echoes. It turns out that the incoherent echoes generated in this way can be much larger than the echoes generated with coherent light.<sup>39</sup> This experiment led to investigations involving time delayed mixing with incoherent light.<sup>30,32</sup> These experiments in turn showed us there was a need for a theoretical analysis which went beyond perturbation theory. We developed such a theory<sup>41</sup> and are now applying it in order to understand problems associated with the interaction of noisy light and matter.

Our increased understanding of the interaction of light and matter led us to use coherent lasers in conjunction with time delayed four wave mixing, which in turn enabled us to develop a new technique which we called Ultrafast Modulation Spectroscopy. 35,36 This technique involved a time delayed four wave mixing experiment using coherent lasers configured to produce picosecond beats in Na vapor. This technique is Doppler free. A more recent advance involves a variant of this technique which produces attosecond beats. 42

We have begun to use the time delayed four wave mixing technique to study relaxation in glasses doped with CdS and CdSe. We have been able to observe relaxation processes in the tens of femtosecond regime. The technique is promising. The time resolution only depends on the bandwidth of the laser noise. We are also setting up to look at ultrafast relaxation processes in GaAs quantum wells.

#### PUBLICATIONS OF WORK SUPPORTED BY ONR 1978 - 1988 Contract #N00014-78-C-0517

- 1. A. Flusberg, R. Kachru, T. Mossberg, and S. R. Hartmann, "Foreign-Gas-Induced Relaxation of Rydberg S and D States in Atomic Sodium," Phys. Rev. A 19, 1607 (1979).
- 2. T. W. Mossberg, R. Kachru, E. Whittaker, and S. R. Hartmann, "Temporally Recurrent Spatial Ordering of Atomic Population in Gases: Grating Echoes", Phys. Rev. Lett. <u>43</u>, 851. (1979)
- 3. T. Mossberg, A. Flusberg, R. Kachru, and S. R. Hartmann, "Total Scattering Cross Section for Na on He Measured by Stimulated Photon Echoes," Phys. Rev. Lett. 42, 1665 (1979).
- 4. R. Kachru, T. W. Mossberg, and S. R. Hartmann, "Stimulated Photon Echo Study of Na(32S1/2)-CO Velocity-Changing Colision," Opt. Comm. 30, 57 (1979).
- 5. R. Kachru, T. W. Mossberg, E. Whittaker, and S. R. Hartmann, "Optical Echoes Generated by Standing-Wave Fields: Observations in Atomic Vapors," Opt. Comm. 31, 223 (1979).

- 6. T. W. Mossberg, R. Kachru, and S. R. Hartmann, "Observation of Quantum-Mechanical Velocity-Changing Collisions," Phys. Rev. Lett. 44, 73 (1980).
- 7. R. Kachru, T. W. Mossberg, and S. R. Hartmann, "Noble-Gas-Induced Broadening of Rydberg S and D States in Atomic Sodium," Phys. Rev. A 21, 1124 (1980).

- S. R. Kachru, T. W. Mossberg, and S. R. Hartmann, "Noble-Gas Broadening of the Sodium D Lines Measured by Photon Echoes," J. Phys. B: Atom. Molec. Phys. 13, L363 (1980).
- 9. R. Kachru, T. W. Mossberg, and S. R. Hartmann, "N2-and CO-induced Collisional Broadening of Forbidden Ground-to-Rydberg-State Transitions in Sodium as Measured by Trilevel Echoes," Phys. Rev. A 22, 1962 (1980).
- 10. T. W. Mossberg, E. Whittaker, R. Kachru, and S. R. Hartmann, "Noble-gas-induced Collisional Broadening of the  $3P_{1/2}$ - $3P_{3/2}$  Transition of Sodium Measured by the Trilevel-Echo Technique, Phys. Rev. A 22, 1962 (1980).
- 11. T. W. Mossbesrg and S. R. Hartmann, "A Diagrammatic Representation of Photon Echoes and Other Laser-Induced Ordering Processes in Gases," Phys. Rev. A 23, 1271 (1981).
- 12. J. H. Eberly, S. R. Hartmann, and A. Szabo, "Propagation Narrowing in the Transmission of a Light Pulse Through a Spectral Hole," Phys. Rev. A 23, 2502 (1981).
- 13. R. Kachru, T. J. Chen, S. R. Hartmann, and P. R. Berman, "Measurement of a Total Atomic-Radiator-Perturber Scattering Cross Section," Phys. Rev. Lett. <u>47</u>, 902 (1981).
- 14. T. W. Mossberg and S. R. Hartmann, "Coherent Transients Theorem: A Comment," Physical Review A 24, 2247 (1981).
- 15. R. Beach, S. R. Hartmann, and R. Friedberg, "Billiard Ball Echo Model," Proceedings of the Int. Cong. on Lasers '81, 991 (1981).
- 16. R. Kachru, T. J. Chen, T. W. Mossberg, and S. R. Hartmann, "Relative Noble-Gas-Induced Broadening of the D lines of Atomic Lithium," Phys. Rev. A <u>. 25</u>, 1546 (1982).
- 17. K. P. Leung, T. W. Mossberg, and S. R. Hartmann, "Noble-gas-induced Collisional Broadening of the 6P<sub>1/2</sub>-6P<sub>3/2</sub> Transition of T1 Measured by Raman Echoes," Phys. Rev. A <u>25</u>, 3097 (1982).
- 18. R. Beach, S. R. Hartmann, and R. Friedberg, "The Billiard Ball Echo Model," Phys. Rev. A 25, 2658 (1982).

- 19. P. R. Berman, T. W. Mossberg, and S. R. Hartmann, "Collision Kernels and Laser Spectroscopy," Phys. Rev. A <u>25</u>, 2550 (1982).
- 20. K. P. Leung, T. W. Mossberg, and S. R. Hartmann, "Observation and Density Dependence of the Raman Echo in Atomic Thallium Vapor," Opt. Comm. 43, 145 (1982).
- 21. T. W. Mossberg, R. Kachru, T. J. Chen, S. R. Hartmann, and P. R. Berman, "Noble Gas Induced Relaxation of the Li 3S-3P Transition Spanning the Short Term Impact Regime to the Long Term Asymptotic Regime," published in the Laser Spectroscopic Conference, Jasper, Canada (1982).
- 22. R. Beach, B. Brody, and S. R. Hartmann, "Photon Echoes in Lithium Vapor with the Use of Angled Excitation Beams," Phys. Rev. A <u>27</u>, 2925 (1983).
- 23. R. Beach, B. Brody, and S. R. Hartmann, "Elliptical Billiard-ball Echo Model," Phys. Rev. A <u>27</u>, 2537 (1983).
- 24. S. R. Hartmann, "Application of the Billiard Ball Model to Problems in Atomic Physics," Proceedings of the Third Symposium on Laser and Applications, Kanpur, India, December 17-21, 1983.
- 25. R. Beach, B. Brody, and S. R. Hartmann, "Photon Echoes with Angled Beams," <u>Laser Spectroscopy VI</u>, Proceedings of the Sixth International Conference, Interlaken, Switzerland, 1983, ed., H. P. Weber and W. Luthy (Springer-Verlag, Berlin, 1983), Vol. 40, p. 48.
- 26. R. Beach, B. Brody, and S. R. Hartmann, "Photon Echoes Made Simple," Laser Chem. 2, 3 (1983).
- 27. R. Beach, B. Brody, and S. R. Hartmann, "Angled-beam Photon Echoes," J. Opt. Soc. Am. B 1, 189 (1984).
- 28. R. Beach, B. Brody, and S. R. Hartmann, "Photon Echoes and Elliptical Billiard Balls," Coherence and Quantum Optics V, Edited by L. Mandel and E. Wolf, Plenum Publishing Corporation, 1984.
- 29. R. Beach and S. R. Hartmann, "Incoherent Photon Echoes," Phys. Rev. Lett. 53, 663 (1984).

- 30. R. Beach, D. DeBeer and S. R. Hartmann, "Time Delayed Four Wave Mixing Using Intense Incoherent Light," Phys. Rev. A <u>32</u>, 6, 3467 (1985).
- 31. Chen Tian-Jie and S. R. Hartmann, "The Response of a Tri-Level Atomic System to Laser Pulses and Two-Photon Tri-Level Echo," Acta Physica Sinica 34, 1034 (1985).
- 32. D. DeBeer, L. G. Van Wagenen, R. Beach and S. R. Hartmann, "Time-Delayed Four-Wave Mixing in Sodium Vapor," Proceedings of the First International Laser Science Conference, Dallas, Texas, 1985, ed., William C. Stwalley and Marshall Lapp, (American Institute of Physics, New York, 1986), p. 592.
- 33. T. J. Chen, D. DeBeer, and S. R. Hartmann, "Observation and Relaxation of the Two-Photon-Excited-State Trilevel Echo in Sodium Vapor," Proceedings of the First International Laser Science Conference, Dallas, Texas, 1985, ed., William C. Stwalley and Marshall Lapp, (American Institute of Physics, New York, 1986), p. 437.
- 34. E. Y. Xu, F. Moshary and S. R. Hartmann, "Noble Gas Induced Collisional Line Broadening of Atomic Li Rydberg States nS and nD (n = 4 to 30) by Trilevel Echoes," Proceedings of the First International Laser Science Conference, Dallas, Texas, 1985, ed., William C. Stwalley and Marshall Lapp, (American Institute of Physics, New York, 1986), p. 439.
- 35. R. Beach, D. DeBeer, L. G. Van Wagenen and S. R. Hartmann, "Picosecond Modulation Spectroscopy in Sodium Vapor," Proceedings of the Fritz Haber International Symposium on Methods of Laser Spectroscopy, Israel 1985, ed., Yehiam Prior, Abraham Ben-Reuven and Michael Rosenbluh, (Plenum Press, New York and London, 1986), 87-94.
- 36. D. DeBeer, L. G. Van Wagenen, R. Beach and S. R. Hartmann, "Ultrafast Modulation Spectroscopy," Phys. Rev. Lett. <u>56</u>, 1128 (1986).
- 37. T. J. Chen, D. DeBeer and S. R. Hartmann, "Observation and Relaxation of the Two-Photon-Excited-State Trilevel Echo in Sodium Vapor," JOSA <u>B3</u>, 493, (1986).

- 38. E Y. Xu, F. Moshary and S. R. Hartmann, "Noble Gas Induced Collisional Line Broadening of Atomic Lithium Rydberg Superposition States 2S-nS and Measured By 2S-nD (n=4 to 30) Trilevel Echoes," JOSA <u>B3</u>, 497 (1986).
- 39. S. R. Hartmann, "Using Incoherent Light to Generate Coherent Excitations," Proceedings of the International Laser Science Conference II, Seattle, Washington, 1986; <u>Advances in Laser Science II</u>, eds. W. C. Stwalley and M. Lapp, AIP Conference Proceedings 160; Optical Science and Engineering Series No. 8 (AIP New York 1987), 556-562.
- 40. D. DeBeer, S. R. Hartmann and R. Friedberg, "Comment on Diffraction Free Beams," Phys. Rev. Lett. <u>59</u>, 2611 (1987)
- 41. R. Friedberg and S. R. Hartmann, "A Diagrammatic Technique for Calculating Radiation of Coherently or Incoherently Excited Two-level Atoms," J. Phys. B: At, Mol. Opt. Phys. 21, 683 (1988)
- 42. D. DeBeer, E. Usadi, and S. R. Hartmann, "Attosecond Beats in Sodium Vapor," Phys. Rev. Lett. <u>60</u>, 1262 (1988)